Appendix BB
Relating Peak Ozone to Degree of Flow Reversal, Volatile Organic Compound (VOC) Reactivity, Solar Radiation, and NO_x in the Houston-Galveston Area

B. Georgoulias, F. Mercado
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Summary

This analysis examined the relationship between ozone and variables describing volatile organic compound (VOC) reactivity, degree of flow reversal, NO_x, and solar radiation (SR) in the Houston-Galveston area. While many of these variables often shared some relationship with ozone, the strengths of these relationships were dependent upon several factors, such as wind direction during the peak ozone hour and when the most "disorganization" in winds across the domain occurred. This analysis found that these relationships were less definitive for the subset of ozone-exceedance days than when all days in the ozone season were considered. Most of the time, reactivity and flow reversal descriptor variables did not explain as much variance in the peak hourly ozone value as average morning wind speed, maximum hourly NO_2 at 8:00 AM, and solar radiation. These last three variables together explained over 65 percent of the variance in peak 1-hr ozone on all days, but only 17 percent of the variance on exceedance days. However, this work is on-going, with many variations of these indicator variables and relationships yet unexplored. Challenges to this analysis include non-linear relationships and monitoring network bias.

Introduction

The TCEQ previously identified similar wind patterns on many ozone exceedance days during 1998 through 2000 in the Houston area (TCEQ Technical Support Document to HGA SIP Revision, 2002). The most striking feature *on average* for all exceedance days was a "rotational flow" around the city, though the pattern varied on specific days. Other work has focused on VOC reactivity, indicative of the potential for these compounds to form ozone (TCEQ Technical Support Document to HGA SIP Revision, 2002; Carter, 1994). The TCEQ was therefore interested in determining the relationship between ozone and rotational-flow characteristics, reactivity, and other important variables in and around Houston.

Analysis

This analysis examined days in June through September of years 1998 though 2001. The objective was to use multiple linear regression to relate important predictor variables to peak ozone; this investigation was a first step towards a statistical model for high ozone formation.

The first task was to characterize the "extent of flow reversal" and/or "extent of rotation" of the winds in the area. As in previous analyses, the TCEQ examined composite averages of hourly wind data across the Houston-Galveston (HGA) domain. One approach to characterizing rotation was to first "draw concentric circles" out from the origin of the composite average trajectory for each hour, as demonstrated for two hours in **Figure IV-1**. (The smaller circle intersects at around 6:00 AM LST, and the larger intersects at noon; the yellow point represents an ozone exceedance of 127 ppb at the Monroe monitoring site around 2:00 PM on this date.)

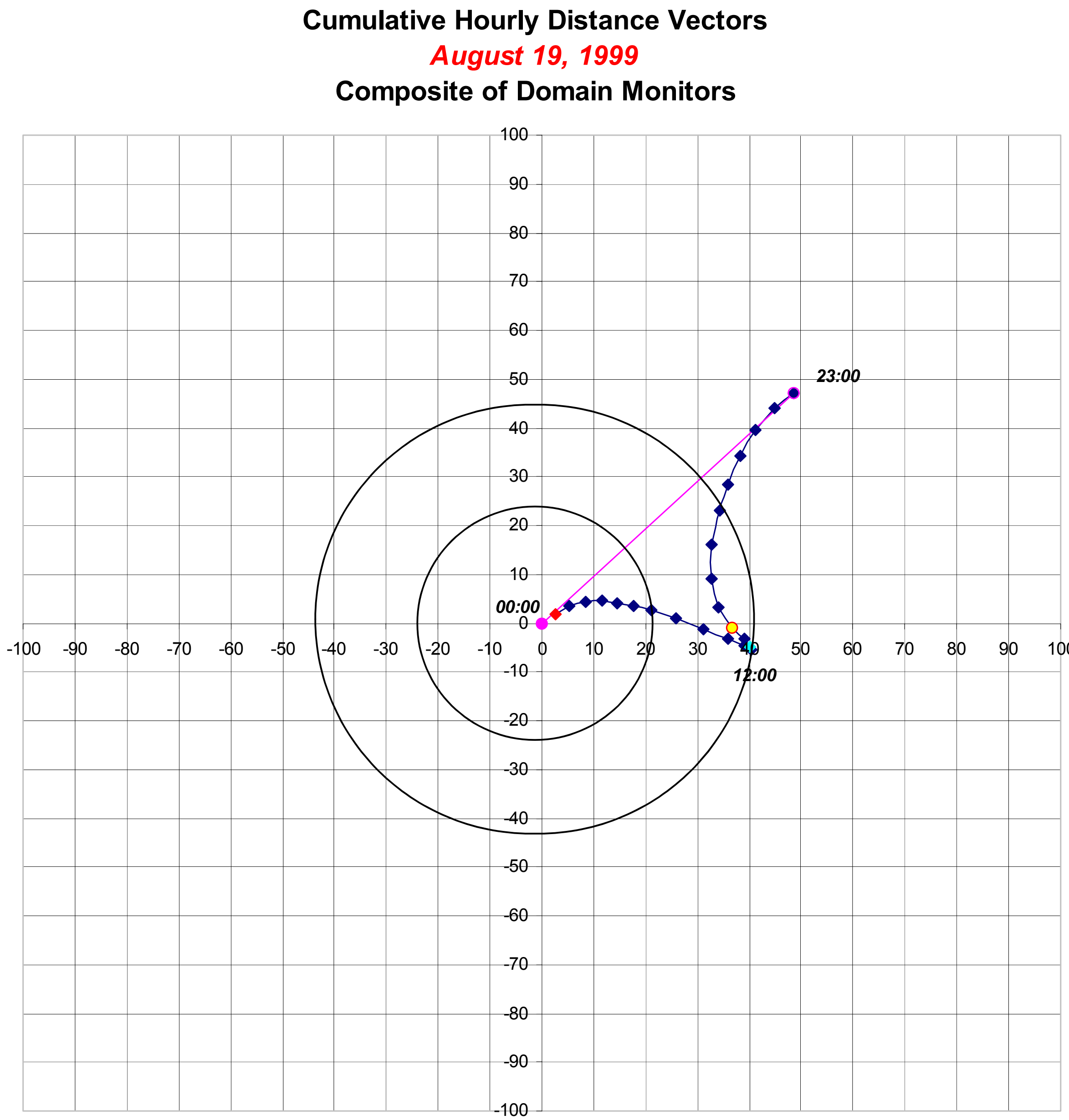

Figure IV-1 – Describing "Extent of Rotation"

A ratio was then calculated for each hour by summing the number of points in the entire day's trajectory that fell within that hour's circle and dividing the sum by the (hour + 1). The larger the ratio above 1.0, the more points from later in the day had passed back into the radius of an earlier hour, and the stronger the likelihood of rotation occurring sometime during the day. Because some hours could have a ratio less than 1.0 (a "tight" rotation will bring points in the trajectory path closer to the origin than earlier hours), the "extent of rotation" value for the day was defined as the sum of ratios greater than 1.0.

Unfortunately, all trajectory rotation patterns do not “turn in on themselves;” some just spiral outward moving further from the origin each hour. Other flow reversal patterns are not necessarily characterized by “rotation;” for example, if the trajectory simply changed direction suddenly at mid-day but did not turn inward closer to the origin, the “extent of rotation” value would not capture that pattern (ratios for all hours would be 1.0). For these variations, another ratio was used to characterize the trajectory pattern–a distance ratio, described by **Equation IV-1**:

$$\text{Distance Ratio: } \frac{\text{Distance traveled by the composite average trajectory in 24 hours}}{\text{Distance of the net vector between Hr 0:00 and Hr 23:00}}$$

The larger the distance ratio, the more the trajectory “meandered” without traveling very far out of the city by the end of the day. In **Figure IV-1**, the magenta line shows the net vector, and the distance traveled is traced by the blue line of the trajectory. Obviously, the distance ratio (DR) will be high for days with a large extent of rotation, too, but the DR also captures flow reversal occurrences. In a sense, the distance ratio is a measure of the “extent of ventilation.”

The TCEQ was also interested in relating VOC reactivity to ozone in the HGA area. To calculate reactivity, we used the Maximum Incremental Reactivity (MIR) scale (Carter, 1994), which describes the maximum ozone formation potential of a VOC. This analysis considered maximum hourly total reactivity (sum of all VOC reactivities) recorded during the 8:00 AM through 12:00 PM hours in the domain. This window was chosen because diurnal patterns showed VOC levels tend to drop significantly in the afternoon (presumably as these compounds become involved in the photochemical reactions that produce ozone).

Other variables are important in the ozone formation process as well. Solar radiation, which drives the reaction between nitrogen dioxide (NO_2) and oxygen (O_2) to form ozone (O_3) (Seinfeld, 1998), is integral to ozone chemistry. Likewise, NO_2 concentrations–particularly in the morning–play a critical role.

This analysis considered daytime peak hourly ozone values (the maximum recorded in the Houston-Galveston domain, which included monitors in Clute and Conroe) from all days in the June-September window for the four years. We also explored relationships when days were grouped by different criteria, such as by ozone exceedance days and non-exceedance days; the hour when winds were “most disorganized” (blowing from the most different directions) across the domain; and the direction of the winds on average at the peak ozone hour. Each grouping changed the significance of some relationships and ability of the variables to explain the variance in the peak ozone value.

Results

Unfortunately, even while “extent of rotation” (sum of rotation ratios > 1.0) and “extent of ventilation” (distance ratio) told us about what the composite trajectory for each day looked like, neither rating was a significant factor in predicting ozone, no matter how days were grouped.

What did prove to be the most telling about the peak ozone value was simply the average morning wind speed (from 8:00 AM through noon). Morning wind speed alone could explain around 35 percent of the variance in peak hourly ozone (adjusted $R^2 = 0.35$), and a linear regression model worked best with the log of both these variables (ln(peak hourly ozone) ~ ln(avg AM wind speed)), $R^2 = 0.36$ (**Figures IV-2 and IV-3**). As expected, peak ozone was higher when wind speeds were lower–when ventilation was low and ozone precursors had a chance to accumulate through the morning.

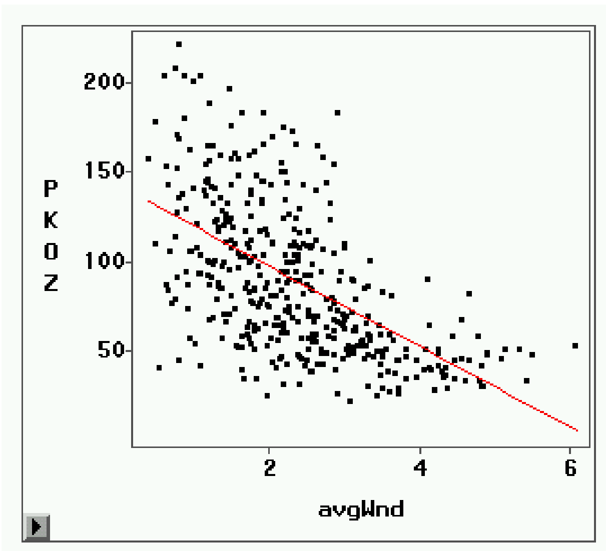

Figure IV-2 – Scatter Plot of Peak 1-hour Ozone (ppb) vs. Morning Average Wind Speed (m/s)

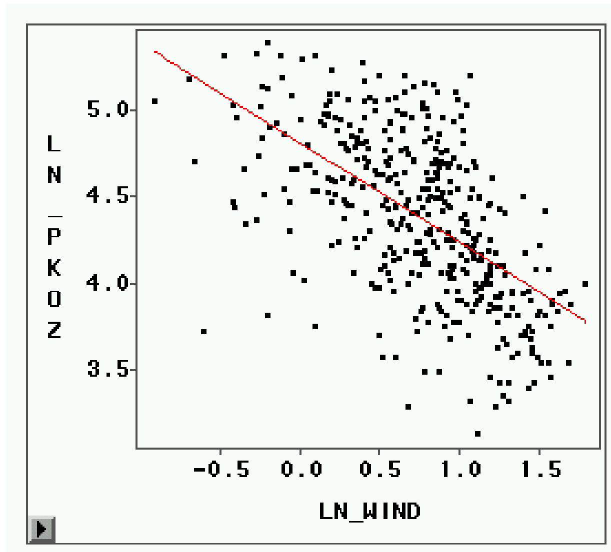

Figure IV-3 – Scatter Plot of ln(Peak 1-hour Ozone, ppb) vs. ln(Morning Ave. Wind Speed, m/s)

Morning NO_2 concentration was also an important predictor of peak ozone. Maximum hourly NO_2 concentration (ppb) at 8:00 AM (across the domain) and morning average wind together predicted about 45 percent of the variance in daily peak hourly ozone (**Table IV-1**). Adding maximum hourly total VOC reactivity recorded between 8:00 AM and noon improved the regression only modestly ($R^2 = 0.47$). (Though note that the coefficient of reactivity is negative, perhaps because of an inverse relationship with wind speed.) Solar radiation (langley/minute) was also important to predicting ozone. Including average solar radiation from 10:00 AM to 2:00 PM (recorded at the Deer Park 2 monitor, which had the most data from 1998 to 2001) with the above variables achieved an R^2 value of 0.66 (See **Table IV-1**); however, this addition rendered VOC reactivity an insignificant variable (at the 0.05 significance level). While this consequence seems surprising because the MIR scale was developed specifically as an indicator of ozone formation potential, there are several possible explanations for this result.

Table 1

Relating Peak 1-hour Ozone to Predictor Variables in HGA Area

N	Equation	RMSE*	Adjusted R^2
455	$\ln(pkhr\_O_3) = 4.53 + 0.013(max\_NO_2) - 0.49[\ln(wind\_speed)]$	0.34	0.45
455	$\ln(pkhr\_O_3) = 4.56 + 0.016(max\_NO_2) - 0.51[\ln(wind\_speed)] - 0.0003(max\_reactivity)$	0.35	0.47
453	$\ln(pkhr\_O_3) = 3.78 + 0.012(max\_NO_2) - 0.55[\ln(wind\_speed)] + 0.86(SR)$	0.28	0.66

*Root Mean Square Error = Standard Deviation of the Error

One possible reason is that ozone's relationship with reactivity is very non-linear, particularly as reaction rates increase and/or become more complex. Some effects of reactivity may also be accounted for in the wind variable–the faster the wind speeds, the less VOC concentration in each "parcel" of air carried away from a source, and the lower the total reactivity in those parcels. The negative coefficient for reactivity in the second equation above is some evidence for this possibility. Another important consideration is monitor network bias; in many cases, plumes of ozone precursors and/or ozone may be narrow or heading in a direction where monitors will not "see" the high ozone that is formed elsewhere in the HGA area. Later in this analysis, we will try to address this issue to some degree by examining relationships for days grouped by wind direction.

Despite total reactivity's poor performance as a predictor in this linear regression approach, morning wind speed, early morning NO_2 concentration, and mid-day solar radiation explained over half the variance in daily peak 1-hour ozone during the months June through September. This is significant, considering the complexity of the chemistry and meteorology at work in the Houston area. The next question was: did these same variables predict daily peak 1-hour ozone on exceedance days and non-exceedance days just as well?

Exceedance Days and Non-Exceedance Days

When the days were divided, there were 93 exceedance days (peak 1-hour ozone > 125 ppb) and 360 non-exceedance days. These three variables could explain about 55 percent of the variance in peak 1-hour ozone on non-exceedance days, but they could only explain about 17 percent of the variance in peak ozone on exceedance days (**Table IV-2**). Also, solar radiation became insignificant in the model on exceedance days.

Table IV-2 – Non-exceedance Days vs. Exceedance Days

Category	N	Equation	RMSE	Adjusted R^2
Non-exceedance Days	360	$\ln(pk\_O_3) = 3.78 + 0.012(max\_NO_2) - 0.55[\ln(wind\_speed)] + 0.86(SR)$	0.25	0.55
Exceedance Days	94	$\ln(pk\_O_3) = 3.78 + 0.012(max\_NO_2) - 0.55[\ln(wind\_speed)] + 0.86(SR)$	0.15	0.17

It is conceivable that solar radiation would contribute significantly to explaining variance in ozone for all days, whereas it might not be as important for concentrations above 125 ppb. By the time ozone has reached such levels, conditions conducive to ozone formation have likely persisted, which means plenty of sunshine. Therefore, the variations in solar radiation on days when 1-hour ozone reaches above125 ppb may not tell us much about *how* high the ozone concentration will go.

VOC reactivity was still not a significant predictor in these relationships. However, this result does not suggest that reactivity is not important to high ozone. We did find that the average maximum hourly reactivity (MIR) in the morning on exceedance days was statistically different (at the 0.05 level) than the average on non-exceedance days. In addition, other work has found the most elevated VOC reactivities coincide with ozone production rates greater than 45 ppb/h, most often in the industrial areas of Houston (Kleinman et al., 2002)–though it should be noted that Kleinman et al.'s work defined reactivity as the lifetime of the OH radical with respect to reaction with a VOC.

Hour When Winds Were "Most Disorganized"

To explore how peak ozone's relationship with these predictor variables might improve under different conditions, this analysis looked more closely at wind data. In animations of wind directions recorded across the HGA monitoring network, something stood out on ozone days; often during the morning, winds appeared to blow from several different directions at the various monitors across Houston (**Figure IV-4**). We wondered if the hour this disorganization occurred could tell us anything about the peak ozone value.

To determine the hour with the most disorganization, we divided the compass grid into 24 bins, each representing a 15° angle slice of the compass. The average wind direction for each hour between 6:00 AM and noon at each monitor was assigned to its respective bin, and the percentage of bins out of the 24-piece "pie" that had winds was calculated for each hour. The larger the percentage, the more winds had blown from varying directions, and the more "disorganization" across the network (for example, the date and hour shown above had a "disorganization score" of 62.5 percent, or wind directions coming from 15 of the 24 bins). Days were then grouped by the first hour with the maximum disorganization.

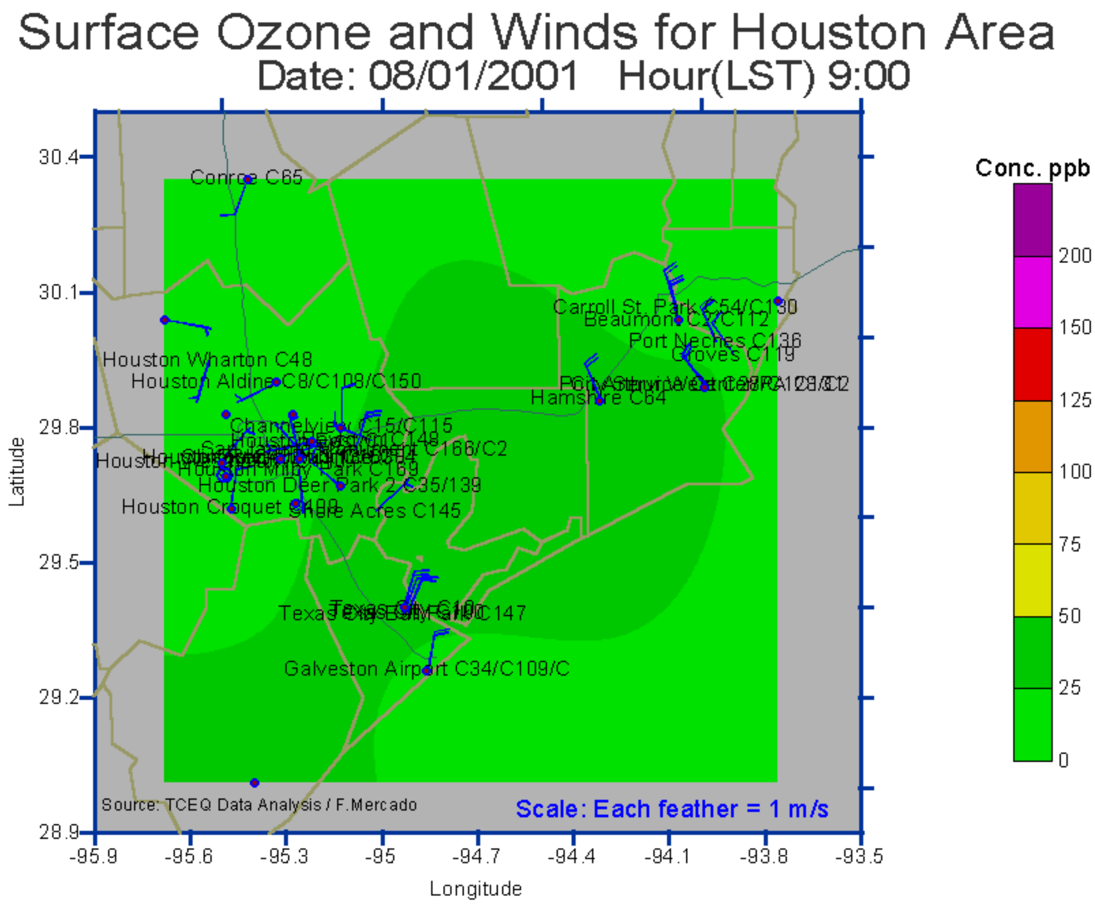

Figure IV-4 – Wind Speed and Directions at 9:00 AM on August 1, 2001 in the Houston-Galveston Domain

For hours 7:00 AM through noon, various combinations of solar radiation, 8 AM NO_2 concentration, and morning wind speed variables explained 65 to 72 percent of the variance in peak ozone for all days; maximum reactivity was still not a significant variable (see **Table IV-3**). Early morning NO_2 concentration was also not a significant predictor until winds fell apart later in the morning–during hours 11:00 AM and noon. Results varied when these regressions were broken into ozone exceedance and non-exceedance days (**Table IV-4**). Most often, the variance in peak ozone above 125 ppb was more difficult to explain with these variables than lower values. The exception appears to be when winds were most disorganized during the 7:00 AM hour; however, there were only 8 exceedance days in that category.

Table IV-3
Relationships Grouped by Hour of Most Disorganization (All Days in June through September, 1998-2001)

First Hour of Most Disorganized Winds	N	Significant Variables related to ln(pk_O_3)	RMSE	Adjusted R^2
6:00 AM	208	ln(wind_speed), SR, max_NO_2	0.26	0.62
7:00 AM	44	ln(wind_speed), SR	0.28	0.72
8:00 AM	20	ln(wind_speed), SR	0.25	0.71
9:00 AM	28	ln(wind_speed), SR	0.26	0.72
10:00 AM	27	ln(wind_speed), SR	0.30	0.67
11:00 AM	57	ln(wind_speed), SR, max_NO_2	0.27	0.65
12:00 PM	69	ln(wind_speed), SR, max_NO_2	0.28	0.71

Table IV-4 – Relationships to Ozone Grouped by Hour of Most Disorganization (Non-Exceedance & Exceedance Days)

	First Hour of Most Disorganized Winds	N	Significant Variables related to ln(pk_O_3)	Adjusted R^2
Non-Exceedance Days	6:00 AM	185	ln(wind_speed), SR, max_NO_2	0.51
	7:00 AM	36	ln(wind_speed), SR, max_NO_2	0.73
	8:00 AM	13	ln(wind_speed)	0.53
	9:00 AM	20	ln(wind_speed), SR, max_reac	0.59
	10:00 AM	19	ln(wind_speed), SR	0.59
	11:00 AM	36	ln(wind_speed), SR	0.41
	12:00 PM	50	ln(wind_speed), SR	0.62

	First Hour of Most Disorganized Winds	N	Significant Variables related to ln(pk_O_3)	Adjusted R^2
Exceedance Days	6:00 AM	23	ln(wind_speed), SR, max_NO_2	0.07
	7:00 AM	8	ln(wind_speed)	0.77
	8:00 AM	7	ln(wind_speed), SR	0
	9:00 AM	8	ln(wind_speed), SR	0
	10:00 AM	8	ln(wind_speed), SR	0
	11:00 AM	20	max_NO_2	0.29
	12:00 PM	19	ln(wind_speed), SR, max_NO_2	0

What is interesting is that different variables appear to be important to predicting ozone, depending on when the winds "fell apart" during the morning hours. For example, when winds were most disorganized late in the morning at 11:00 AM, SR, NO_2, and wind speed were all significant predictors of peak hourly ozone. However, when days were split between exceedance and non-exceedance days for this hour, morning NO_2 alone explained almost 30 percent of the variance on exceedance days, while wind speed and SR explained around 40 percent of the variance in non-exceedance days. Unfortunately, patterns were not consistent (e.g., NO_2 was not always significant on only exceedance days or only when winds were disorganized later in the morning), but this does demonstrate possible differences between the key players in ozone formation on some days and the dependence on meteorology.

Wind Direction

Apparent early in this analysis was the importance of wind data to predicting peak ozone. Not only is the wind speed important in determining how precursor compounds accumulate in the Houston area, but wind direction plays a key role in what the monitoring network "observes." Therefore, it is important to consider peak ozone value grouped by wind direction–but the wind direction when and where?

First, we grouped days by the *composite average* wind direction in the domain at the hour of the peak ozone. **Table IV-5** shows these results grouped by quadrant (SE, NE, NW, SW). All variables except reactivity explained 65 to 71 percent of the variance in peak ozone when the average winds were from the northeast or southeast during the peak ozone hour, but only about 45 percent of the variance when winds were from the southwest. Morning wind speed and SR explained about 74 percent of the variance in peak ozone when winds were generally from the northwest during the peak ozone hour, though there were considerably less days in this category–only 18. When days were split into exceedance and non-exceedance days, variables explained significantly less of the variance on exceedance days than when peak 1-hour ozone was less than 125 ppb (**Table IV-6**). There were not enough exceedance days with winds from the SW or NW to obtain a meaningful regression model.

Table IV-5
Relationships Grouped by Composite Average Wind Direction During Peak Ozone Hour (All Days in June through September, 1998-2001)

Quadrant Containing Wind Direction	N	Significant Variables related to ln(pk_O_3)	RMSE	Adjusted R^2
SE	276	ln(wind_speed), SR, max_NO_2	0.26	0.71
NE	64	ln(wind_speed), SR, max_NO_2	0.25	0.65
SW	94	ln(wind_speed), SR, max_NO_2	0.29	0.45
NW	19	ln(wind_speed), SR	0.27	0.74

Table IV-6
Relationships Grouped by Composite Average Wind Direction During Peak Ozone Hour (Non-Exceedance & Exceedance Days)

	Composite Average Wind Direction	N	Significant Variables related to ln(pk_O_3)	Adjusted R^2
Non-Exc. Days	SE	216	ln(wind_speed), SR, max_NO_2	0.63
	NE	41	ln(wind_speed), SR	0.62
	SW	89	ln(wind_speed), SR, max_NO_2	0.45
	NW	14	ln(wind_speed), SR	0.60
Exceedance Days	SE	60	ln(wind_speed)	0.12
	NE	23	None	0.05
	SW	5	N/A	N/A
	NW	5	N/A	N/A

We also tried assigning direction to the 15^{o} angle bin rather than the compass quadrant, but not many bins had very many days. Another problem with the groupings above is that each quadrant may be influenced by which monitors recorded the peak ozone. Also, while composite average wind direction gives us an idea of what was happening in the domain overall, it is not necessarily consistent with the wind direction at the monitor where the peak ozone value was recorded. To examine these issues, our next step was to group days by wind direction recorded at the peak ozone monitor and by monitor site.

Results were not necessarily better with monitor-specific wind direction than before with composite average wind direction (**Table IV-7**). One difference was that maximum morning reactivity became a significant variable, though its coefficient was negative. There were some interesting results when exceedance and non-exceedance days were subdivided. For ozone exceedance days, only two quadrants had enough days to apply multiple linear regression: when winds were from the SE and NE direction at the monitor. As observed previously, relationships were not as strong on exceedance days as when all days were considered together. When winds at the ozone monitor during the exceedance days were from the SE, morning wind speed alone explained just under 20 percent of the variance in the peak ozone value. When winds were from the NE on exceedance days, it appeared that NO_2 at 8:00 AM and maximum morning reactivity could explain over 35 percent of the variance in peak ozone value–the highest percentage thus far (but with only 18 days) and one of the few times reactivity was a significant predictor variable (but again, its coefficient was negative). By contrast, when winds were from the same direction on non-exceedance days, SR and wind speed were the significant variables, explaining nearly twice as much variance ($R^2 = 0.67$). When peak ozone hour winds were from the SE on non-exceedance days, all four variables were significant (**Table IV-7**).

Table IV-7
Relationships Grouped by Average Wind Direction
During Peak Ozone Hour at the Ozone Monitor

	Average Wind Direction at Ozone Monitor	**N**	**Significant Variables related to ln(pk_O_3)**	**Adjusted R^2**
All Days	SE	238	ln(wind_speed), SR, max_NO_2, max_reactivity	0.71
	NE	59	ln(wind_speed), SR	0.69
	SW	69	ln(wind_speed), SR	0.55
	NW	13	ln(wind_speed)	0.53
Non-Exc. Days	SE	184	ln(wind_speed), SR, max_NO_2, max_reactivity	0.58
	NE	41	ln(wind_speed), SR	0.67
	SW	64	ln(wind_speed), SR	0.60
	NW	12	None	0.51
Exceedance Days	SE	54	ln(wind_speed)	0.19
	NE	18	max_NO_2, max_reactivity	0.36
	SW	5	N/A	N/A
	NW	0	N/A	N/A

One reason for the differences could be the distribution of the monitors recording the peak ozone. This is important information because where monitors are located could have a large influence on what predictor variables were correlated more strongly with the peak ozone recorded there, along with where the winds were blowing. For example, a frequency distribution of monitors revealed which sites weighted more heavily in the SE group (**Table IV-8**).

Table IV-8 – Frequency Counts of Monitors with SE Winds During Peak 1-Hour Ozone (Records That Include All Four Regression Variables)

AIRS ID	Site Name	Days	Percent of Total Days	No. Exceedance Days
48-039-1003	Clute	4	1.7	4
48-167-0014	Galveston Airport	7	2.9	4
48-167-1002	Texas City C10	1	0.4	0
48-201-0024	Aldine	64	26.9	10
48-201-0029	NW Harris	42	17.6	4
48-201-0051	Croquet	10	4.2	7
48-201-0055	Bayland Park	9	3.8	7
48-201-0066	Westhollow	4	1.7	1
48-201-0070	Houston Regional Office	1	0.4	1
48-201-1034	Houston East	10	4.2	3
48-201-1035	Clinton	4	1.7	3
48-201-1039	Deer Park 2	9	3.8	5
48-201-1050	Seabrook Friendship Park	3	1.3	2
48-339-0089	Conroe	70	29.4	3

On non-exceedance days, the Conroe site (AIRS 48-339-0089) north of Houston recorded the most one-hour peak ozone values (70-3 = 67 days). Therefore, it was useful to examine results subdivided by AIRS site to see if relationships stood out for particular monitors. **Table IV-9** shows the results for the Conroe site. For non-exceedance days at Conroe when winds were from the SE during the peak ozone hour, the same three variables achieved an R^2 of 0.69, and for all days, 0.74. Too few exceedance days were available for the Conroe site to perform a meaningful regression analysis.

Table IV-9
Relationships at the Conroe Monitor (48-339-0089)
Grouped by Wind Direction During Peak Ozone

Quadrant Containing Wind Direction	N	Significant Variables related to ln(pk_O_3)	RMSE	Adjusted R^2
SE	70	ln(wind_speed), SR, max_NO_2	0.17	0.74
SW	12	ln(wind_speed)	0.13	0.81

Grouping by AIRS monitor *and* wind direction usually restricted the number of days to too few, especially when considering exceedance days only. In cases where there were enough days at a monitor to be grouped by wind direction, the significant variables were not always the same as when all sites were considered together. While such results might be helpful in distinguishing ozone behavior in different locations around the Houston area, it also demonstrates the difficulty of characterizing relationships between ozone and predictor variables across the HGA domain.

Variations on Chosen Variables

There were several different ways to define the variables discussed above; in an effort to refine some of the relationships, we tried a few variants in this analysis. When some diurnal patterns showed high VOCs before 8:00 AM in the morning, total VOC reactivity during an earlier time frame (from 3:00 to 8:00 AM) seemed like it might be a better choice. However, this change did not improve regression models appreciably overall. In addition, if the 8:00 AM NO_2 concentration was taken from the same AIRS station as peak VOC reactivity, results did not improve significantly.

Conclusion

This analysis found average morning wind speed, average mid-day solar radiation, and maximum NO_2 concentration at 8:00 AM to be important predictor variables of peak ozone concentration in the Houston-Galveston area domain. Together, these variables could explain over 65 percent of the variance in peak 1-hr ozone for all days in the June through September time frame, in years 1998-2001. Parameters designed to characterize "extent of rotation" were not nearly as helpful as morning wind speed, which was the variable with the most influence on peak hourly ozone in this analysis.

Future Work

Another useful approach might be to examine days with rapid ozone formation (i.e., a large change in ozone concentration in the span of an hour or two). We will also investigate multiplying VOC reactivities by wind speed to "normalize" VOC concentrations and further explore the relationship of reactivity with ozone.

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